Review Paper:

Removal of Synthetic Dyes from Textile Wastewater using Physical and Biological Methods: A Review

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Abstract

Dyes are a significant group of organic pollutants that are well-known for harming both humans and aquatic life in general. Dye-contaminated wastewater needs to undergo appropriate treatment before being released into main streams in order to minimize its detrimental impact on people and the environment. Adsorption, flocculation-coagulation, reverse osmosis and biological treatment are some current technologies that are frequently used to remove dye from wastewater.

Keywords: Dyes, Textile industry, Adsorption, Reverse osmosis, Biological method.

Introduction

Water, a valuable and necessary component of life, makes up around seventy-one percent of the earth's surface; 97.5 percent of it is salt water, 2.5 percent is fresh water and 0.007 percent is suitable for drinking. Due to the improper release of used water from industry into the environment, contamination of water is currently one of the biggest issues affecting the entire world. There are many chemical industries that work with dyes, but the textile industries alone use a significant amount of dye and discharge wastewater as a result of the process. In textile factories, both dry and wet processes are used to produce fibre[7].

The wet process generates a significant amount of effluent that is heavily contaminated and uses a lot of water, chemicals, auxiliary chemicals, dyes. Sizing, de-sizing, sourcing, bleaching, mercerizing, dyeing, printing and finishing techniques are used in the wet process[32].

Textile processing technology

The following procedures make up the standard textile processing technology:

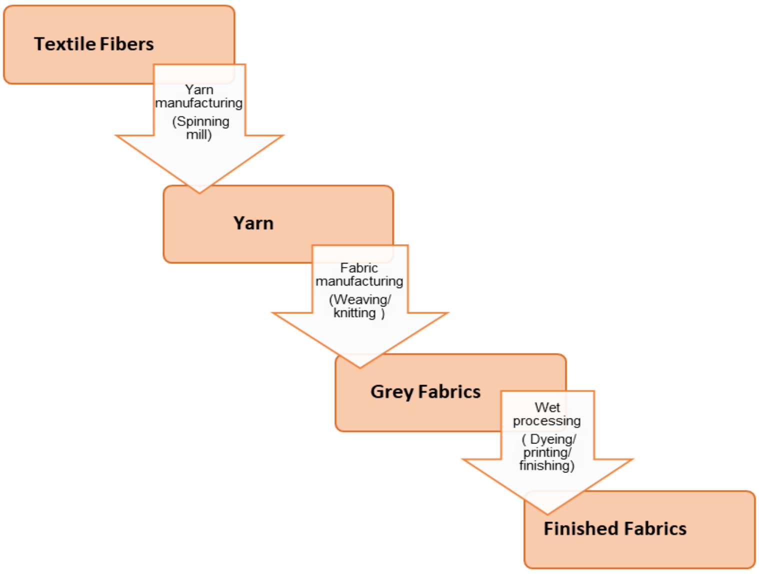


Figure 1: Flowchart of textile processing[7]

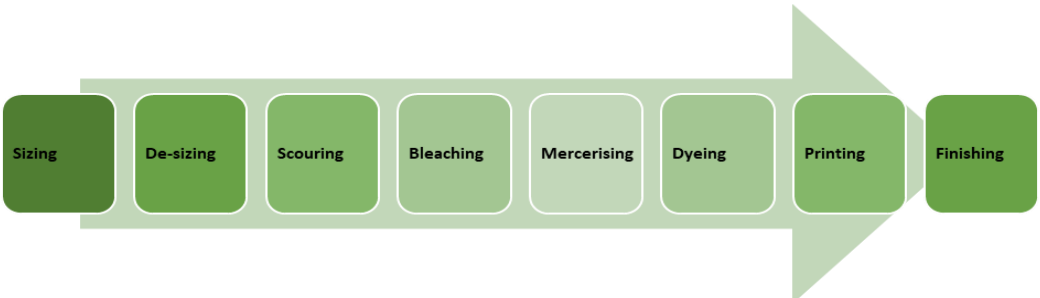


Figure 2: Flowchart of textile wet processing[32]

- Sizing is the first phase in the procedure and it comprises the use of sizing chemicals such as starch, polyvinyl alcohol (PVA) and carboxy methyl cellulose to provide strength to the fibres and to reduce breaking.
- Desizing is the method used to take sizing agents out of the fabric before weaving. Wastewater from this procedure contains enzymes, starch and waxes.
- Scouring is the procedure whereby impurities are eliminated from the fibres by treating them with an alkali solution, usually sodium hydroxide, to dissolve natural oils, fats, waxes and surfactants.
- Bleaching is the phase when undesirable colour is removed from the fibres using chemicals like hydrogen peroxide and sodium hypochlorite.
- The stage before dyeing is known as "mercerising" which involves treating the fabric with a concentrated alkaline solution and washing it in an acidic solution. Mercerising improves the fiber's dyeability, shine and attractiveness.
- The process of adding colour to the fibres is known as dyeing. It often requires a lot of water both during the dye bath and the rinse process. Numerous chemicals are used to enhance dye adsorption onto the fibres including metals, salts, surfactants, organic processing aids, sulphide and formaldehyde.
- Printing is the application of colour to cloth in predetermined patterns or designs.
- Finishing is the last step in the textile manufacturing chain that enhances the fabrics' aesthetic appeal[23].

Dyes

Being one of the fundamental components of nature, colour enhances the aesthetic value and fascination of human life. Currently, a variety of chemicals are employed as colouring agents. They can be classified into two main categories: dyes and pigments which are soluble and insoluble in water respectively. Dyes can be characterised as compounds that impart colour without being impacted by elements such as light, water and surfactants[33].

Components of dye: Dyes comprise of two basic components such as chromophore and auxochrome. Chromophores are responsible for colour because of their ability to absorb light in the UV region. The ethylene, azo, carbonyl, sulphur group, nitro and nitroso groups are the most significant groups of chromophores. Auxochrome, on the other hand, enhances the casting of the chromophore by altering the overall strength of the electronic system and giving dye solubility and adhesion to the fibre via stable chemical bonds. The carboxyl, sulfonate, amino and hydroxyl groups are the most common auxochromes[25]. Dyes are frequently used in paper, leather, fur, hair, pharmaceuticals, cosmetics, waxes, greases, plastics and textile industry[12].

In the past, natural dyes were used to colour the fabric. However, the variety of colours produced by these was limited and dull. Furthermore, when washed and exposed to sunshine, they had poor colour fastness. They therefore need a mordant to create a dye complex that would fix the fibre and dye together. Since W. H. Perkins' discovery of synthetic dyes in 1856, a broad variety of dyes has been developed that are colorfast, have a larger colour spectrum and have brighter hues. As a result, "dye application" has grown significantly over time[14]. According to the Color Index, more than 10,000 different types of dyes are currently produced globally, resulting in 700,000 tons of dye output.

Due to their simplicity of synthesis, low cost, firmness, great stability to light, temperature, detergent and microbial attack, as well as their wide range of colour options when compared to natural dyes, synthetic dyes are being employed more and more in the textile and dyeing industries. Additionally, extremely polluted effluents have been released as a result of this. An average dye concentration of 300 mg/L has been found in textile manufacturing process effluents which is higher than the normal threshold of 1 mg/L at which colour becomes perceptible[6].

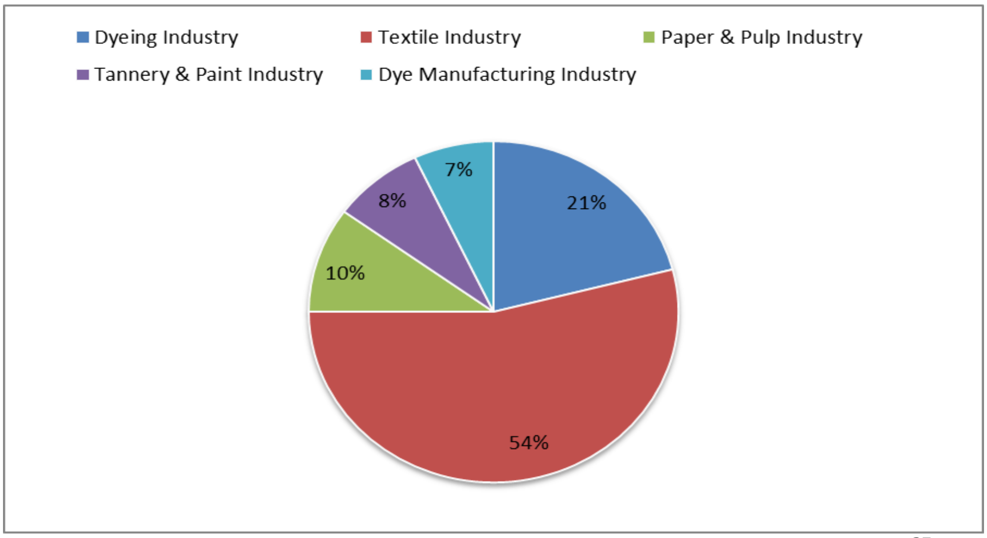


Figure 3: Comparison of dye effluent emission from different industries[27]

Classification of dyes
Dyes can be classified in many ways but here we are classifying them on following basis:
1. On the basis of their industrial applications (Table 1)
2. On the basis of chemical structure (Table 2)
3. On the basis of general structure (Table 3)

The chemical structure classification system is the most suited system for dye classification. This approach offers a lot of benefits. First, it easily recognises dyes as belonging to a group with distinct features such as azo dyes (strong and cost-effective) and anthraquinone dyes (weak and expensive). Second, it is the classification that both synthetic dye chemists and dye technologists utilise most frequently. Thus, terms like an azo yellow, an anthraquinone red and a phthalocyanine blue are easily recognised by both chemists and technicians[28]. Depending on their general structural characteristics, the various dyes used in the textile industry can be divided into cationic, non-ionic and anionic dyes.

Effects of dyes: One of the biggest and most significant industries in the world is the colouring industry. However, due to the range of colours included in its wastewater, it is thought to be an environmentally damaging source of contamination for organisms. Heavy metals like iron, lead, nickel, copper, zinc and chromium are present in trace concentrations in textile dyeing effluents. These dyeing effluents are released into the nearby waterway, agricultural fields, irrigation channels and outside water where they eventually end up in water bodies like rivers, the sea etc.

These dyeing effluents may cause variation in the physical, chemical and biological nature of the aquatic atmosphere by the nonstop alteration in turbidity, odor, noise, temperature, pH etc. which are injurious to community health, livestock, wildlife, fish and biodiversity. The colloidal matter present along with colors and oily scum increases the turbidity and gives the water a bad appearance and foul smell. It prevents the penetration of sunlight necessary for the process of photosynthesis. This interferes with the oxygen transfer mechanism at the air-water interface. Depletion of dissolved oxygen in water is the most serious effect of textile waste as dissolved oxygen is very essential for marine life. This also hinders with self-purification process of water.

Table 1
Classification of dyes on the basis of their industrial applications[5]

S.N.	Dye class	Industrial applications
1.	Azo	Textile, Printing and Paper manufacturing industry
2.	Basic	Paper, Textile and Plastic industry
3.	Acid	Leather, Foodstuff and Textile industry
4.	Reactive	Textile and Ink printing industry
5.	Disperse	Textile and Plastic industry
6.	Direct	Leather, paper and Textile industry
7.	Sulfur	Textile and Paper industry
8.	Vat	Textile and Leather industry

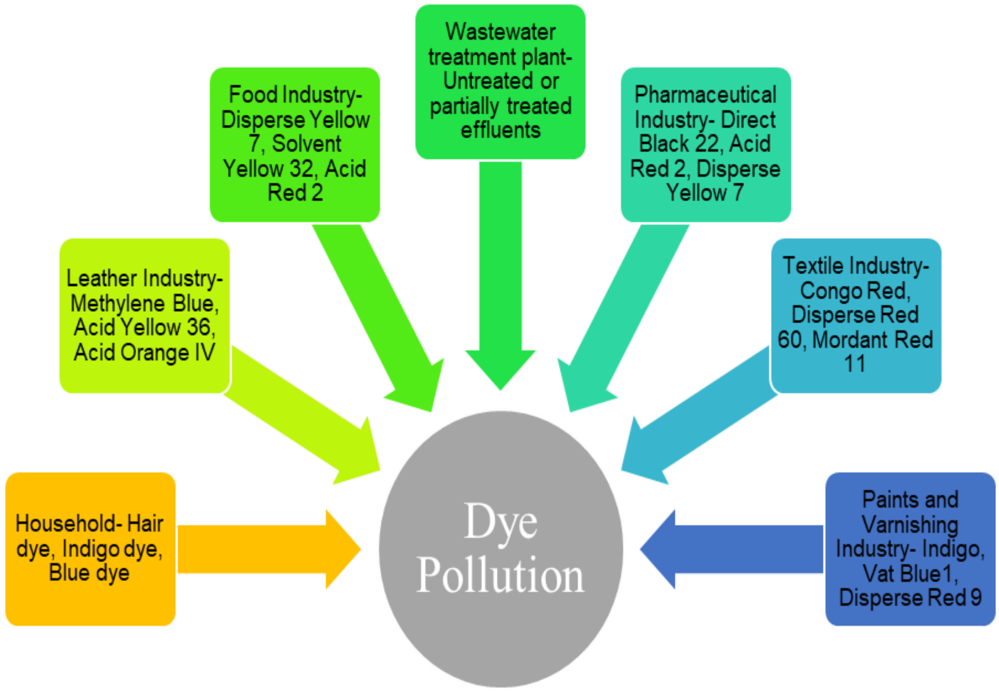


Figure 4: Different sources of dye pollution[8]

Table 2
Classifications of dyes on the basis of chemical structure[11]

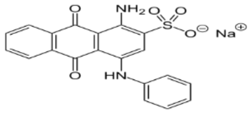
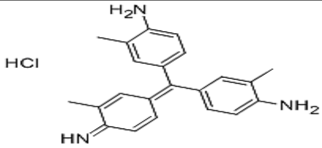
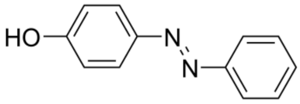
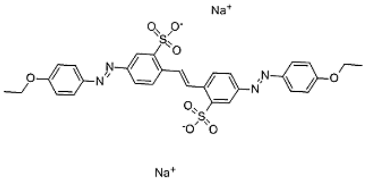
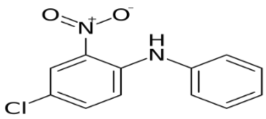
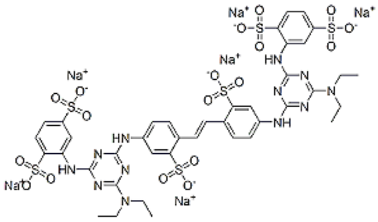
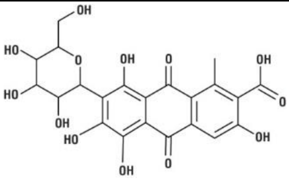
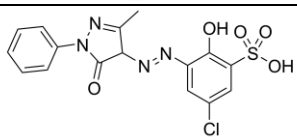
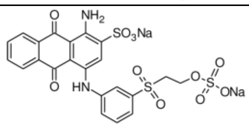
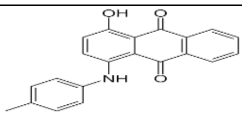
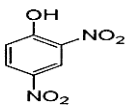
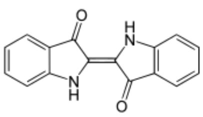
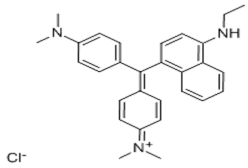
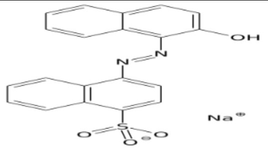
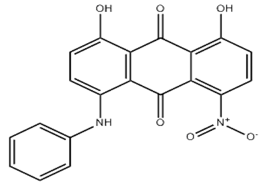
S.N.	Dye class	Chemical structure	General Formula
1.	Acid		$C_{20}H_{13}N_2NaO_5S$ (Acid Blue 25)
2	Basic		$C_{22}H_{24}ClN_3$ (Basic Violet 2)
3.	Azoic components and compositions		$C_{12}H_{10}N_2O$ (Solvent Yellow 7)
4.	Direct		$C_{30}H_{28}N_4NaO_8S^{2+}$ (Direct Yellow 12)
5.	Disperse		$C_{12}H_9ClN_2O_2$ (Disperse Yellow 26)
6.	Fluorescent brighteners		$\mathbf{C_{40}H_{38}N_{12}Na_6O_{18}S_6}$ **(**Fluorescent Brightener 263)
7.	Food, drug and cosmetic		$C_{22}H_{20}O_{13}$ (Carminic acid)
8.	Mordant		$C_{16}H_{13}ClN_4O_5S$ (Mordant red 19)
9.	Reactive		$C_{22}H_{16}N_2Na_2O_{11}S_3$ (Reactive Blue 19)
10.	Solvent		$C_{21}H_{15}NO_3$ (Solvent Violet 13)
11.	Sulfur		$C_6H_4N_2O_5$ (Sulfur Black 1)
12.	Vat		$C_{17}H_{14}N_2O_2$ (Indigo Blue)

Table 3
General structure-based classification of dyes used in the textile industry

S.N.	Dye class	Structure	Types of dyes
1.	Cationic dyes		Basic dyes
2.	Anionic dyes		Acid dyes Reactive dyes Azo dyes Direct dyes
3.	Non-ionic dyes		Disperse dyes

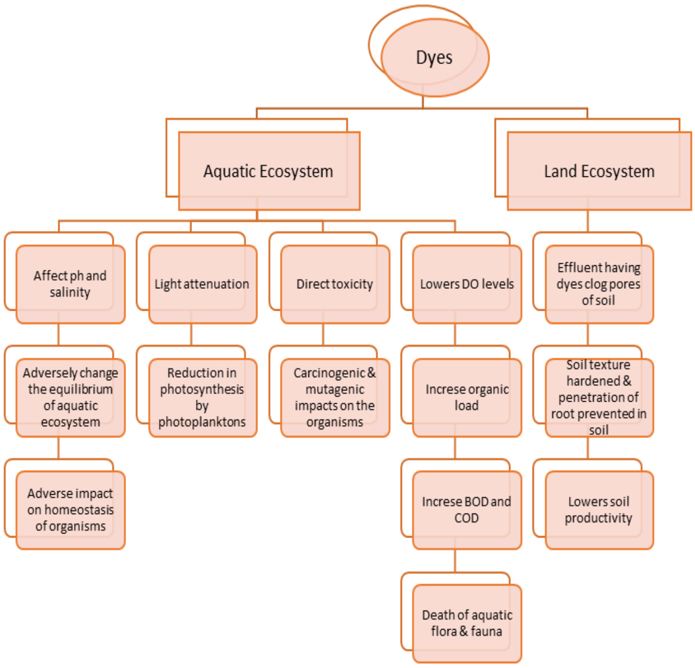


Figure 5: Effects of dyes on aquatic and land ecosystems[26]

In addition, when this effluent is allowed to flow in the fields, it clogs the pores of the soil resulting in a loss of soil productivity. The texture of soil gets hardened and penetration of roots is prevented. The presence of dyes in surface and subsurface water is making them not only appealingly intolerable but also sources of many water-borne diseases viz. mucous membrane, dermatitis, perforation of the nasal septum and severe irritation of the respiratory tract. Adulteration to this aquatic system carries severe hazards to the inclusive epidemic and socio-economic outline inside[12].

Synthetic azo dyes are carcinogenic and toxic posturing a severe health risk to human health. It is also responsible for dysfunction of the kidney, liver, brain; reproductive system and central nervous system are some effects of dyes on human beings[15]. Chemicals evaporate into the air we breathe or are absorbed through our skin and show up as allergic reactions and may cause harm to children even before birth[14]. Due to the high intensity of colors in the water, dyes are highly visible to the human eye even in a very small concentration which is highly unacceptable on aesthetic grounds.

Different methods for removing dyes from textile wastewater: Textile industries use different dyes as per the colour requirement. Some of the examples of dyes are: Methylene blue, Crystal violet, Acid yellow 36, Basic blue 9, Direct yellow, Reactive black and Acid red 119. Most current techniques used for the removal of dyes fall under three main classes -

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Different methods to remove synthetic dyes [25]

Physical methods:
- Adsorption
- Reverse osmosis
- Coagulation/ Flocculation

Biological methods:
- Microorganism
  - Bacteria
  - Fungi
- Enzymes

Chemical methods:
- Oxidation
- Ozonation
- Electrolysis

Physical methods: Physical techniques are adsorption, reverse osmosis, coagulation and flocculation. Due to their effective ability to remove colour and low operating expenses, these techniques are frequently employed in industry[10].

(1) Adsorption: The accumulation of a substance at the interface between two phases is referred to as adsorption. Adsorbent is the solid on which adsorption takes place while adsorbate is the substance that accumulates at the interface. Adsorbent surface area, adsorbent to adsorbate ratio, adsorbent particle size, temperature, pH and contact time are the factors that affect adsorption efficiency[31]. The techniques for treating dye wastewater use a variety of adsorbents including activated carbon, coal, fly ash, silica, wood, clay, agricultural wastes and cotton waste.

The experimental procedure for the adsorption of Methylene blue dye by activated clay (Table 4) is described as follows:
(1) Prepare a 1-L solution with a constant strength of $NaNO_3$ (5×10^{-2} M) and different MB concentrations;
(2) distribute 100 mL of solution to a series of 125-mL polyethylene (PE) bottles;
(3) adjust initial pH to cover a range from 2 to 10 by either HCl or NaOH;
(4) add a given amount of air-dried SAC (0.2 g/L) into the solution;
(5) shake these bottles on a reciprocal shaker at 150 excursions/min for 5 h at 25 ◦C. This contact time was found to be adequate for reaching equilibrium adsorption based on the results of kinetic study;
(6) at the end of shaking, record the final pH of the mixed liquor;
(7) filter the liquor through a 0.45-m filter paper to collect the supernatant and
(8) determine the residual MB concentration in the supernatant[29].

2) Reverse osmosis: Reverse osmosis is characterized by a membrane pore size in the range of 0.5nm. The operating pressures in RO are generally between 7-100 bars. The ability of RO membrane to remove both organic and inorganic compounds has made it attractive for the treatment of textile wastewater[2]. This process gives a high efficiency in the separation of dyes. The product rate in RO increases with increasing temperature, but it decreases with increasing feed concentration and operating time[3].

Table 4
Adsorption capacities of various adsorbents for removal of different dyes

S.N.	Adsorbent	Dye	% removal
1.	Activated clay[29]	Methylene Blue	60-95%
2.	Kaolin[20]	Crystal Violet	65-95%
3.	Activated rice husk[31]	Acid Yellow 36	45-80%
4.	Fly ash[17]	Methylene Blue	36-45%
5.	Sugarcane bagasse[34]	Basic Blue 9	55.5-94%

Table 5
Removal Efficiency of different dyes by Reverse Osmosis

S.N.	Dye	% removal	Technology
1.	Acid Blue[9]	98%	RO membrane
2.	Direct Blue 6[3]	98.89%	RO polyamide membrane
3.	Direct Yellow[3]	98.30%	RO polyamide membrane
4.	Acid Red[2]	97.2%	RO membrane
5.	Reactive Black[2]	99.58%	RO membrane
6.	Reactive Blue[2]	99.9%	RO membrane

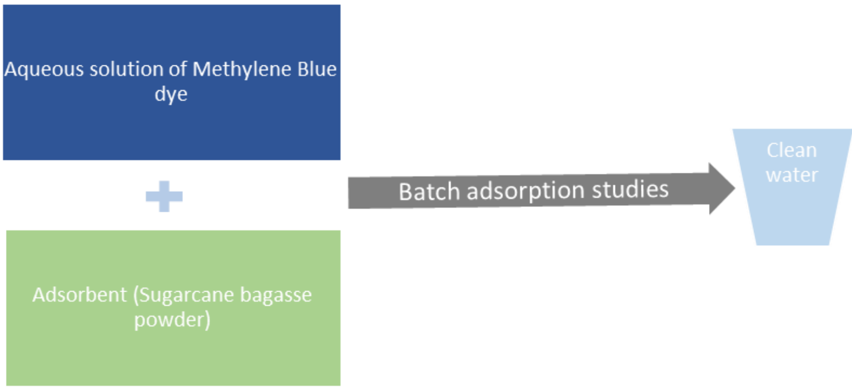
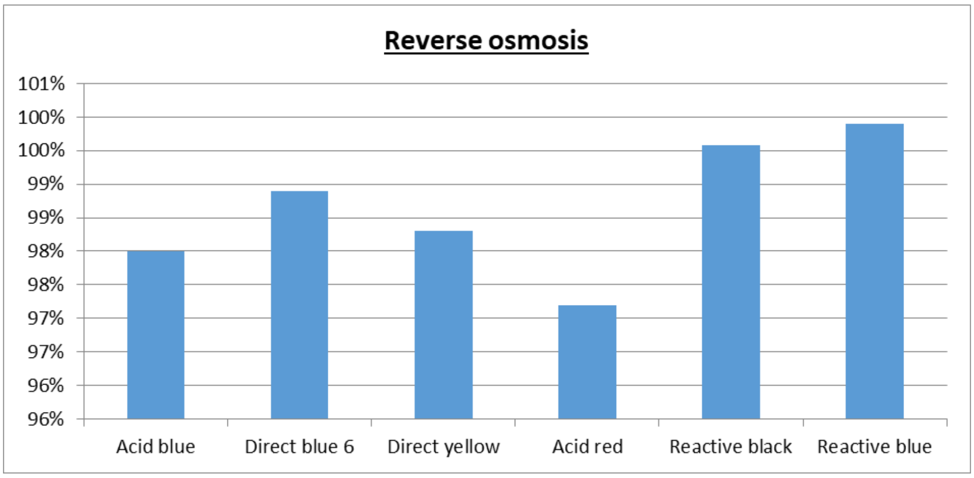


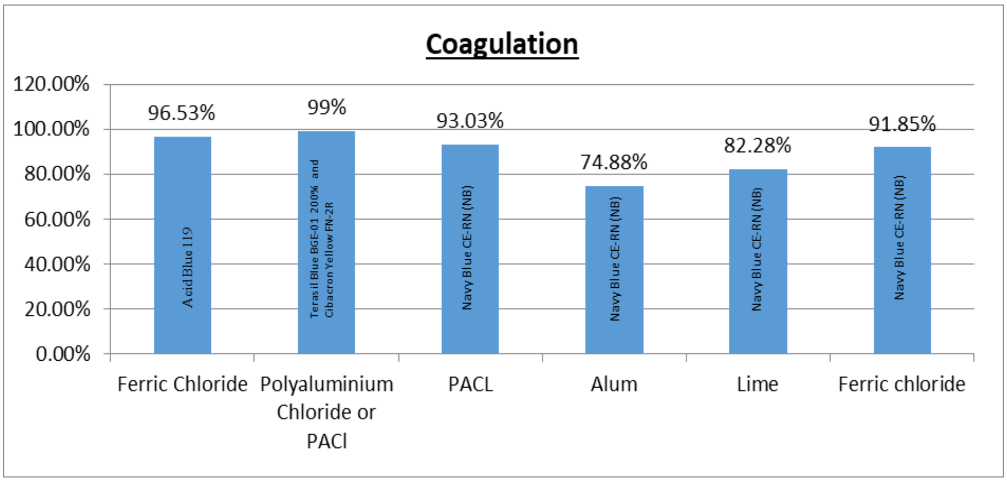
Figure 6: Removal of Methylene Blue (Basic blue 9) by sugarcane bagasse [16]



Graph 1: Comparative removal of different dyes by Reverse Osmosis

Table 6
Different coagulants and % removal of dyes

S.N.	Dye	Coagulant	% removal
1.	Acid Red 119[18]	Ferric chloride	96.53%
2.	Terasil Blue BGE-01 200% and Cibacron Yellow FN-2R[30]	Polyaluminium chloride (PACl)	>99%
3.	Navy Blue CE-RN (NB)[21]	Polyaluminium chloride (PACl)	93.03%
4.	Navy Blue CE-RN (NB)[21]	Alum	74.88%
5.	Navy Blue CE-RN (NB)[21]	Lime	82.28%
6.	Navy Blue CE-RN (NB)[21]	Ferric Chloride	91.85%



Graph 2: Dye removal efficiency of different coagulants

(3) Coagulation-flocculation: Coagulation and flocculation are two separate procedures used in the treatment of wastewater from the textile industry. Flocculation allows the particles to bind together and grow larger so that they may be more easily separated from the water, coagulation allows the particles charges to be neutralised. Both organic and inorganic coagulants are utilised in the treatment of wastewater.

The fundamental advantage of the coagulation-flocculation process is that it may decolorize textile wastewater by removing dye molecules from dye bath effluents rather than by partial degradation of dyes. The efficiency of the coagulation-flocculation process is determined by the pH and temperature of the solution, the type and dose of coagulants and the intensity and duration of mixing[30].

Comparison of Physical methods: Adsorption process provides an attractive alternative for the treatment of polluted waters, especially if the sorbent is inexpensive and does not require an additional pre-treatment step before its application. Reverse osmosis processes can simultaneously remove hardness, color, many kinds of bacteria and viruses and organic contaminants such as agricultural chemicals and tri-halo-methane precursors[2]. Molecules having acidic functional groups which can coordinate with iron to form relatively hydrophobic complexes, can be removed by coagulation. Hence coagulation cannot treat all kinds of dyes.

Cationic dyes cannot coagulate at all, making their removal by this technique impossible. Acid, direct, vat, mordant and reactive dyes usually coagulate, but the resulting floc is of poor quality and does not settle well even after the introduction of a flocculent. Sulfur and disperse dyes coagulate well and settle easily. In the presence of surfactants, the dosing of chemicals has to be significantly increased to achieve satisfactory color removal[13].

Biological methods

Toxic dyes have been successfully removed by biosorption using bacteria, yeast, filamentous fungi and algae. Biosorption is the word used to describe the uptake or accumulation of substances by microbial mass. This property of microorganism is due to the cell wall components such as hetero-polysaccharides and lipids that make up their cell walls which contain a variety of functional groups such as amino, hydroxyl, carboxyl, phosphate and other charged groups. These functional groups bind to the azo dye with strong attractive forces.

Azo dyes are xenobiotic and the use of microbial or enzymatic treatment methods for the complete decolorization and degradation of dyes from textile effluent has a number of advantages over physico-chemical processes including:

- being environmentally friendly;
- being cost-competitive;
- producing less sludge;
- yielding end products that are non-toxic or have complete mineralization and
- requiring less water consumption.

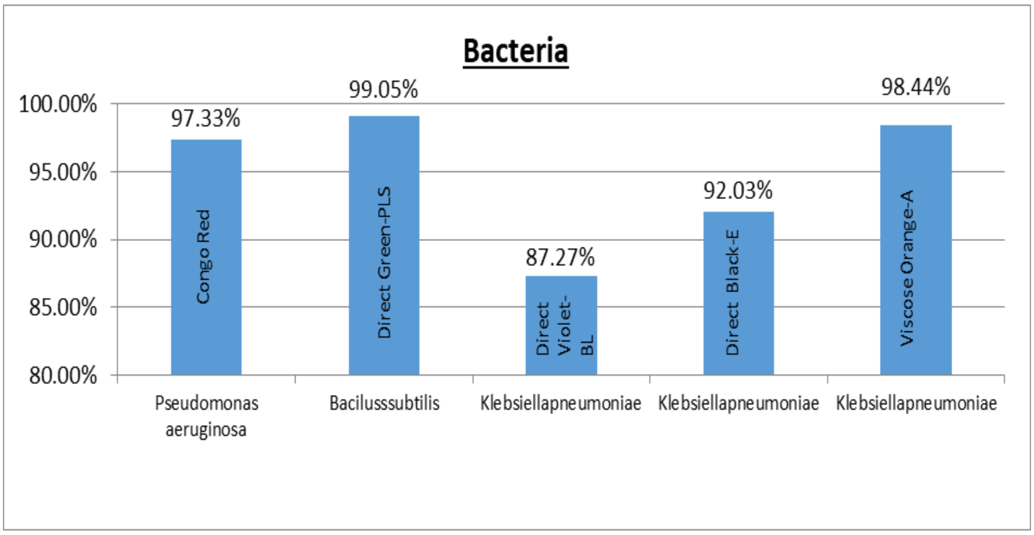
The effectiveness of microbial decolorization depends on the adaptability and activity of the selected microorganisms. Consequently, a large number of species have been tested for the decolorization and mineralization of various dyes in recent years[25].

(1) Bacteria: Several biotechnological approaches have attracted interest concerning degradation of dyes in an eco-efficient manner, mainly with the use of bacteria and often in combination with physicochemical processes. The reduction of the azo dye is usually non-specific and decolorization is faster during the bacterial degradation process. A wide range of aerobic and anaerobic bacteria such as *Pseudomonas aeruginosa*, *Bacillus subtilis*, *Klebsiella pneumoniae*, *Escherichia coli*, *Klebsiella pneumoniae* and *Bacillus halodurans* MTCC 865 have been extensively reported for resulting good biodegradation of azo dyes.

Pseudomonas aeruginosa is widely used in a decolorization study because of its capacity to degrade a variety of dyes (Congo Red, Methyl Red and Tartrazine) and is also exploited in degradation of commercial azo dye used in textile wastewaters. Laccase enzyme found in the *Pseudomonas aeruginosa*, employs the mechanism of free radical that is nonspecific for executing the degradation of the dyes of azo without forming toxic aromatic amines.

Table 7
Removal of different dyes by different bacteria

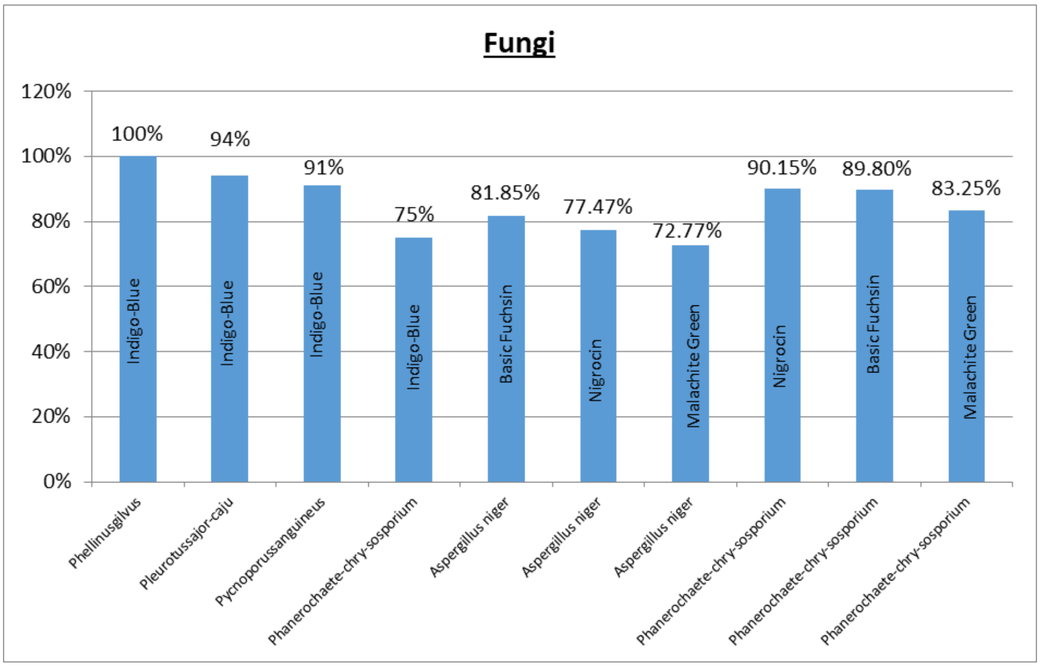
S. N.	Bacteria	Enzyme that helps to degrade dye	Dye	% Removal
1.	*Pseudomonas aeruginosa*[24]	Laccase	Congo Red	97.33%
2.	*Bacillus subtilis*[24]	Azoreductase	Direct Green-PLS	99.05%
3.	*Klebsiella pneumoniae*[24]	Azoreductase	Direct Violet-BL	87.27%
5.	*Klebsiella pneumoniae*[24]	Azoreductase	Direct Black-E	92.03%
6.	*Klebsiella pneumoniae*[24]	Azoreductase	Viscose Orange-A	98.44%



Graph 3: Dye removal efficiency of different bacteria

Table 8
Removal of different dyes by different fungi

S. N.	Fungi	Dye	% removal
1.	*Phellinus gilvus*[4]	Indigo-Blue dye	100%
2.	*Pleurotus sajor-caju*[4]	Indigo-Blue dye	94%
3.	*Pycnoporus sanguineus*[4]	Indigo-Blue dye	91%
4.	*Phanerochaete-chrysosporium*[4]	Indigo-Blue dye	75%
5.	*Aspergillus niger*[22]	Basic Fuchsin	81.85%
6.	*Aspergillus niger*[22]	Nigrocin	77.47%
7.	*Aspergillus niger*[22]	Malachite Green	72.77%
8.	*Phanerochaete-chrysosporium*[22]	Nigrocin	90.15%
9.	*Phanerochaete-chrysosporium*[22]	Basic Fuchsin	89.8%
10.	*Phanerochaete-chrysosporium*[22]	Malachite Green	83.25%



Graph 4: Dye removal efficiency of different fungi

(2) Fungi: Many studies have been reported (Table 7) showing the effective use of the fungi against dyes degradation by producing the azoreductase enzyme for decolorization. Fungi can degrade textile dye effluents through their action or by enzyme digestion produced by them[19]. According to several studies, fungi species including *Phellinus gilvus*, *Pleurotus sajorcaju*, *Pycnoporus sanguineus*, *Phanerochaete chrysosporium* and *Aspergillus niger* are efficient at breaking down dyes.

Comparison of Biological methods: The use of biological methods to remove the color from textile effluents is usually a cheaper alternative since it presents no major processing costs. However, this method presents some disadvantages, since several dyes are designed to resist microbial attack. Biological methods involve the use of bacteria[1]. fungi and algae. Bacterial degradation has been mainly applied in the removal of azo dyes. However, this biological method has been found ineffective in removing color from several dyes. The azo dyes generally resist aerobic degradation. However, its degradation was observed in anaerobic conditions, but aromatic amines are formed as the final product, which despite having no color can be toxic, mutagenic, or carcinogenic.

Enzymes have several features that make them more viable than conventional catalysts: they are biodegradable catalysts, allow the operation at low and high substrate concentrations, allow the operation over a wide range of pH, temperature and salinity, have a reduced sludge formation and are simple and easy to control. All these advantages added to their high specificities and catalytic activities, with the possibility of designing enzymes with the exact desired properties through genetic engineering and computational design, suggest the potential application of this process in the treatment of effluents. Despite the advantages of enzymatic wastewater treatment, the major limitation in the use of enzymes is their prohibitive cost[19].

Conclusion

Dyes are a major class of pollutants that have a significant impact on both general human life and aquatic life. Water containing dye must be carefully handled before being released into the environment to lessen its harmful effects on humans and the environment. Adsorption, reverse osmosis, flocculation-coagulation and biological treatment are examples of common procedures for dye removal. Adsorption and biological treatment are the most frequently investigated techniques over the past few years. Coagulation/flocculation is also a good process if it is used with another process. Indeed, the newer techniques are bringing about several improvements. Most of the techniques can achieve more than 80% dye removal and several exceed 90%.

Great advances in the removal of dyes from wastewater have been reported during the last few years and it is quite encouraging that several reported methods are very fast and have low costs with exciting dye removal efficiencies. Therefore, it is advised that more research should be carried out in this direction because water is the second most precious abiotic component of our ecosystems and its safe treatment and conservation is the duty of every person living on this planet. We hope that more sophisticated technologies are developed so that wastewater can be treated easily with low costs at both industrial and pilot scales.

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